URANIUM DISTRIBUTION IN SEDIMENTS OF THE LOWER SAN PEDRO VALLEY, SOUTH-EAST ARIZONA, AND IMPLICATIONS FOR INDOOR RADON

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INTRODUCTION

Radon is a colorless, odorless gas produced by the natural radioactive decay of uranium. The U.S. Environmental Protection Agency has determined that exposure to indoor radon may increase a person's risk of developing lung cancer. Indoor-radon levels generally correlate with uranium concentration in underlying rocks and soil, and some areas of Arizona are known to have elevated levels of uranium.

This study is part of an ongoing evaluation of potential radon hazards in Arizona by the Arizona Geological Survey and the Arizona Radiation Regulatory Agency. This report presents results of a study of the distribution of uranium in late-Cenozoic basin-fill sediments of the lower San Pedro Valley in southeast Arizona.

Uranium concentrations were measured using a portable gamma-ray spectrometer. The spectrometer survey followed the methods outlined in Duncan and Spencer (1993) and the same equipment was used during both investigations. Indoor-radon data were supplied by the Arizona Radiation Regulatory Agency and well water-radon data were taken from Duncan et al.(1993).

LOCATION

The San Pedro basin is a large, elongate valley in southeast Arizona, extending from northern Sonora, Mexico north-northwest for more than 150 miles to Kelvin, Arizona. The valley is drained by the San Pedro River, a permanent stream, which flows north to Winkelman, where it joins the Gila River, which flows northwest to Kelvin.

This study focuses on the lower portion of the San Pedro basin, defined here as that part of the valley north of Redington, to the northern extent of the valley near Kelvin (Figure 1). The lower San Pedro Valley includes the towns and settlements of Redington, San Manuel, Mammoth, Winkelman, Dudleyville, Hayden, Kearny, and Kelvin.

GEOLOGY

Lower San Pedro Basin

The San Pedro basin is a major structural trough, 2 to 10 miles wide and over 120 miles long in Arizona (the basin continues into Sonora, Mexico), formed during the late-Tertiary Basin and Range disturbance. The lower basin is bordered on the east and northeast by the Galiuro and Dripping Spring Mountains, and on the west by the Santa Catalina-Rincon and Tortilla Mountains. An extensive variety of rock types are found in the bordering mountains, including Precambrian metamorphic and granitic rocks, Paleozoic and Mesozoic sedimentary rocks, and early- to mid-Tertiary volcanic and plutonic rocks.

A thick sequence of late Miocene-Pliocene sediments, known as the Quiburis Formation, was deposited in the lower San Pedro basin. The sediments consist of silt, sand, and gravel interfingered with lacustrine deposits. Silt, clay, gypsum, marl, and diatomite comprise the thick lacustrine section. Erosion since the Pleistocene has formed several terraces in the San Pedro Valley and has exposed several hundred feet of sediments.

Gravity modelling indicates that basin-fill sediments may be nearly 2000 feet thick near Mammoth and 3200 to 4800 feet thick in the Redington area (Oppenheimer, 1980). Well logs indicate at least 1100 feet of lacustrine sediments below the surface along the valley center near Mammoth (Page, 1963).

Previous Studies

The Quiburis Formation was first described by Blake (1902) and was further studied by Heindl

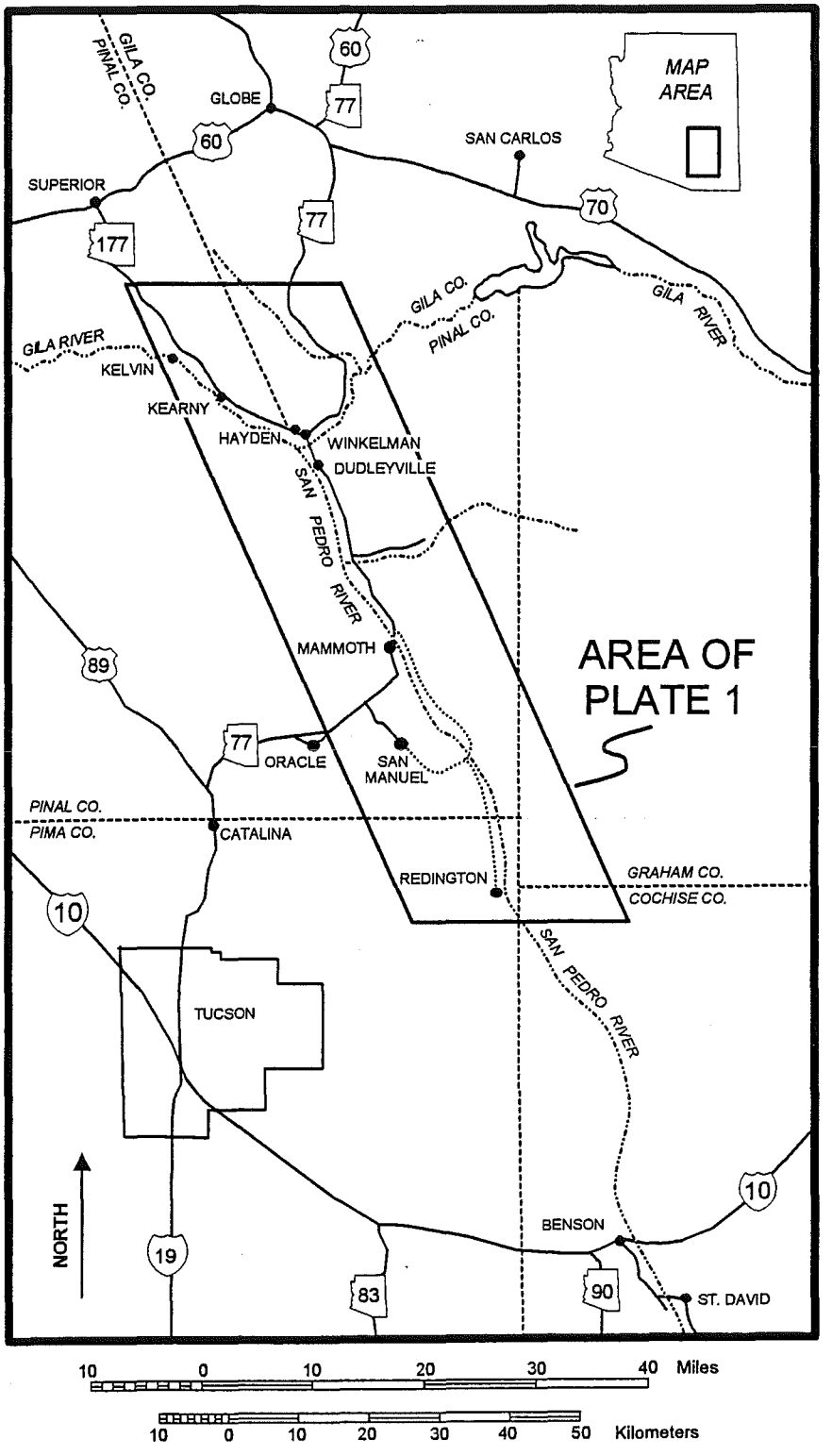

Figure 1. Location of study area. Outlined area shows coverage of plate 1.

(1963) and Creasey (1967), who both considered it to be a subdivision of the more widely distributed Gila Conglomerate. (The term Gila Conglomerate is no longer used in the San Pedro area.) Diatomite deposits near Mammoth were studied in detail by Shenk (1990), and gypsum deposits south of Winkelman are described in Hardas (1966). Other graduate student theses and dissertations on the sediments of the lower San Pedro include Smith (1967), Agenbroad (1967), Ladd (1975), Scarborough (1975), and Utley (1980). Uranium occurrences and favorability were surveyed by the U.S. Atomic Energy Commission (AEC, 1970; Texas Instruments, 1978), and Scarborough (1981).

Mapping by the U.S. Geological Survey includes Heindl (1963), Creasey (1967), Krieger (1968a; 1968b; 1968c; 1974a; 1974b), Banks and Krieger (1974), and Cornwall and Krieger (1975). Dickinson (1991; 1992) detailed the structural history and setting of the San Pedro region.

GAMMA-RAY SPECTROMETER SURVEY

Methods

The Arizona Geological Survey conducted a survey of the lower San Pedro basin using an EG&G geoMetrics model GR-310 portable gamma-ray spectrometer. The machine employs an external detector containing a 347-cm^3, thallium-doped, sodium iodide crystal and a high-gain photomultiplier tube. Four independent channels provide measurements of the diagnostic gamma radiation for uranium (via bismuth-214, 1.76 million electron volts [MeV]), thorium (via thallium-208, 2.62 MeV), which is needed for uranium assay corrections, as well as total gamma radiation (0.4 to 4.0 MeV) and potassium.

Count times of 1, 10, 100, or 1000 seconds may be selected, and due to the random nature of radioactive decay, longer count times generate less statistical error and greater precision. Periodic comparisons of 100- versus 1000-second count times confirmed that the 100-second count time was sufficiently accurate for the purposes of this study and so was used for data acquisition. Uranium concentrations in parts per million (ppm) were calculated from the field data using correction factors and assay equations developed by Duncan and Spencer (1993).

Results

Figure 2 is a histogram of uranium concentrations determined by spectrometer measurements. The mean for 306 assays in the lower San Pedro Valley is 3.65 ppm. Uranium concentrations of 7 ppm or greater are considered anomalous (Duncan and Spencer, 1993). Anomalous concentrations of uranium were measured in sediments near Redington and in the diatomite deposits southeast of Mammoth. Locations of spectrometer measurements and uranium concentrations are shown on Plate 1.

Analyses of 16 samples were performed by a commercial laboratory (Chemex Labs, Inc., Sparks, Nevada), using the delayed neutron counting method, to check the quality of the spectrometer data. Figure 3 shows the results of laboratory analyses versus spectrometer measurements. Ideally, data points would lie along the dashed diagonal line. The points are scattered along a diagonal array around the line, with more below it than above it. This asymmetry suggests that the spectrometer yields slightly higher measurements than the laboratory analyses, but this bias, if real, is minor. All points (except the highest one) fall within a ± 3 ppm U envelope. A major source of the spread of data points away from the 1-to-1, diagonal correlation line, almost certainly results from the fact that the two methods measure different volumes of rock. The spectrometer measures an "average" radiation from a large area (several tens of centimeters to several meters across), which may contain zones of normal as well as anomalous levels of uranium. Gamma rays that reach the detector come from rock near the surface with exponentially decreasing contributions from deeper rocks. Samples collected for chemical analyses, on the other hand, represent a much smaller volume of rock than that sampled by gamma-ray spectrometer measurements. Furthermore, samples collected for chemical analysis are homogenized during sample preparation. Chemical analysis of a collected sample, typically consisting of perhaps a kilogram of rock, may contain

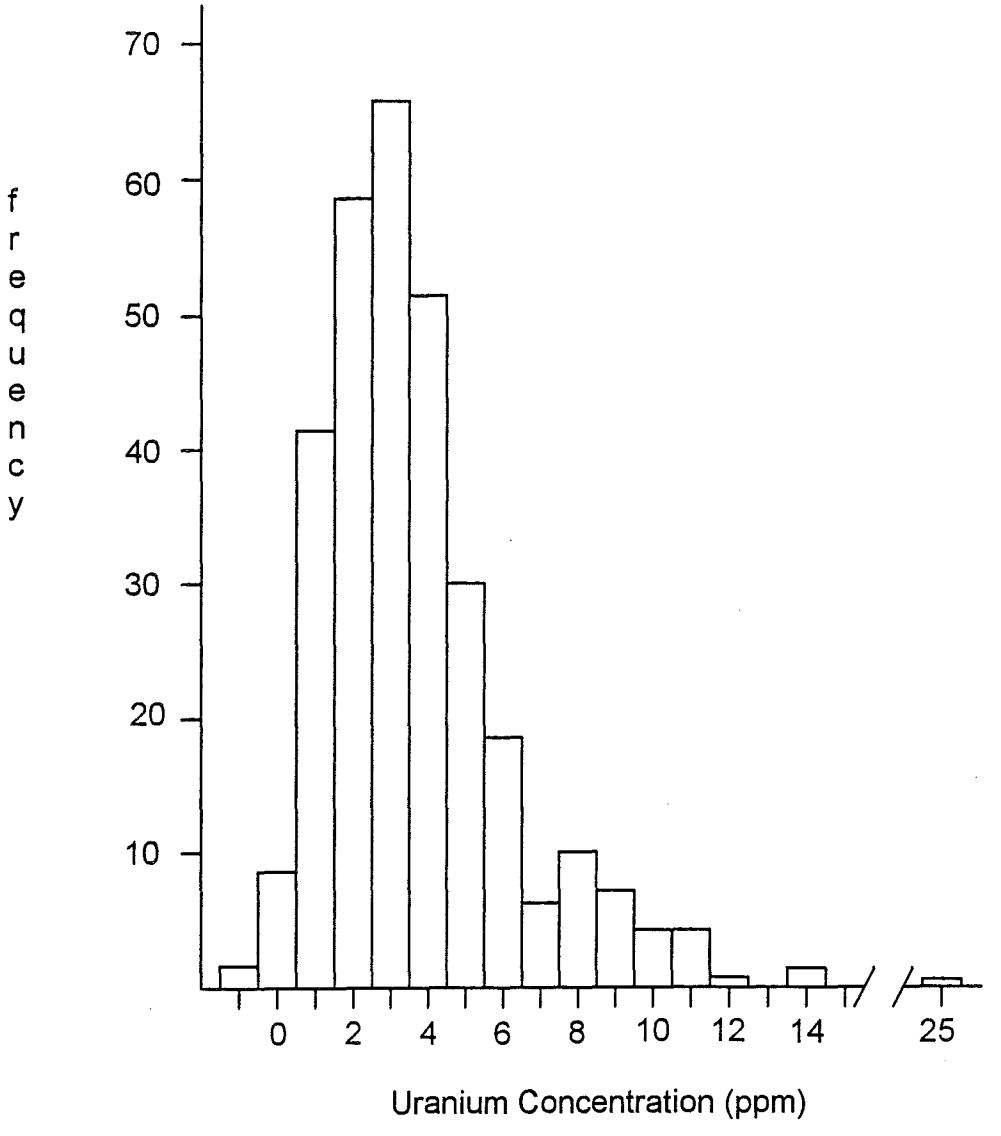

Figure 2. Histogram of uranium concentrations in the lower San Pedro basin. Concentrations greater than 6 ppm are considered anomalous. Values less than zero reflect error introduced during measurement and calculation of concentration (Duncan and Spencer, 1993). and reveal an accuracy of measurement of probably± 2-3 ppm.

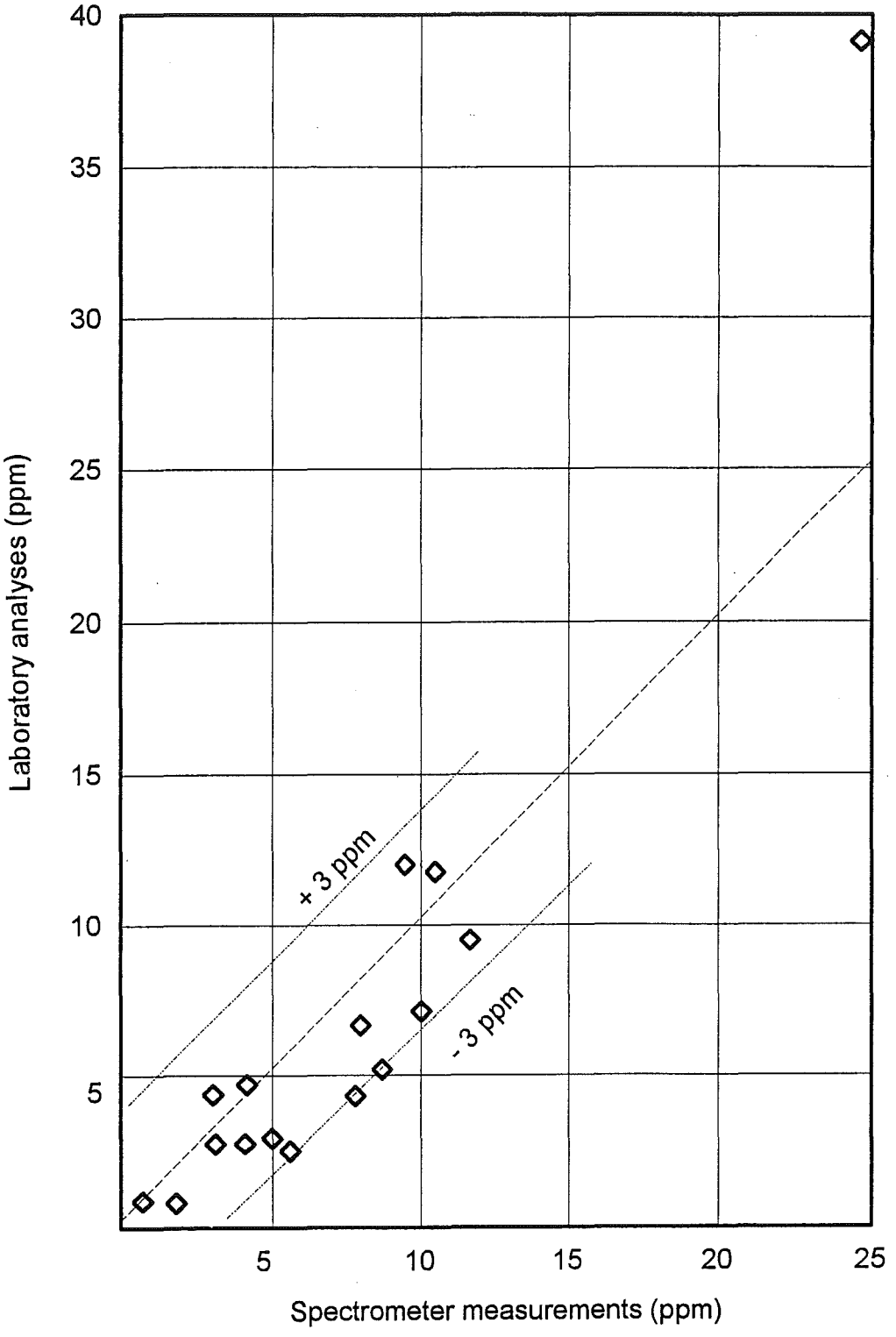

Figure 3. Plot of spectrometer measurements versus commercial laboratory analyses of sediments in the lower San Pedro Valley.

lower or higher uranium levels than the average of the larger surrounding area. Another source of error may be the loss of uranium decay products before bismuth-214 is reached in the uranium-238 decay series. Decay of bismuth-214 is measured by the spectrometer, not actual uranium decay, and uranium content is calculated assuming that loss of decay products between uranium-238 and bismuth-214 has been insignificant. If this assumption is wrong, and intermediate decay products such as radon gas have leaked out in significant quantity, then spectrometer measurements will be systematically lower than laboratory analyses.

URANIUM GEOLOGY

Rock-type Associations

Uranium anomalies in the sediments of late-Cenozoic basins in Arizona are generally restricted to marly, diatomaceous, and cherty lacustrine sediments. These types of uranium-bearing sediments have been described in the Verde Valley (Duncan, 1993) and in the Safford-San Simon and Duncan valleys (Harris, 1994). The basin fill of the lower San Pedro Valley contains abundant marly, diatomaceous and gypsiferous lacustrine sediments. Uranium is concentrated in the marly and diatomaceous sediments, but is virtually absent in gypsum-bearing sediments; sandy to gravelly sediment that forms the remainder of the basin-fill is similarly lacking in uranium.

A U.S. Department of Energy survey (Jones, 1978) generally considered sediments like those found in the San Pedro Valley to be unfavorable for uranium accumulation, largely owing to the oxidized nature of the sediments. However, a U.S. Department of Energy NURE aerial survey (Texas Instruments, Inc., 1979) detected a weak uranium anomaly west and northwest of Redington and this study found scattered anomalous uranium concentrations in the same area. The NURE aerial survey did not detect the anomalous uranium concentrations of the diatomite deposits southeast of Mammoth that were measured in this study.

Uranium concentrations in the bedrock of the surrounding mountain ranges are generally within the range of normal crustal abundances (2 to 4 ppm) with the exception of some impure limestone in the Tertiary Mineta Formation on the northeast flank of the Rincon Mountains, and a few scattered, very small anomalous uranium occurrences in the Dripping Springs and northern Galiuro Mountains (U.S. Atomic Energy Commission, 1970; Scarborough, 1981).

Origin of Uranium Anomalies

The concentration of uranium in basin fill is probably more strongly controlled by the nature of the sediments and depositional environment than the amount of influx of uranium. Much of the sediment exposed in the lower San Pedro Valley was deposited in lacustrine, playa, or paludal (swampy) environments. These conditions promote the deposition of fine-grained sediments and evaporites, including diatomite, gypsum, marl, and organic matter.

Although most carbonate rocks contain very little uranium, especially those deposited in oxidizing environments, some impure carbonates can contain considerably more uranium. Impurities such as clay, organic matter, and silica gel can absorb uranium (Jones, 1978; Schmidt-Collerus, 1979). Tuffaceous sediments may be altered to clay and release silica shortly after deposition, providing sites for uranium adsorption (Zielinski, 1980).

Organic matter in the basin sediments would also contribute to an increased uranium content by reducing the soluble, oxidized form of uranium U(+6) to the insoluble U(+4) state. The presence of organic matter may increase the uranium content by a factor of 10,000 or more over that of the surface runoff or groundwater supplying uranium to the basin (Schmidt-Collerus, 1979).

Conditions during or after deposition of the sediments (at least those presently exposed) in the lower San Pedro basin were apparently favorable for the accumulation or preservation of uranium in the

diatomites, but were unfavorable in the gypsum-bearing sediments. Oxidizing conditions at the time of deposition would have kept uranium in its soluble state and would have prevented the preservation of organic matter which facilitates the precipitation of uranium (Mickle and Mathews, 1978). Oxidizing conditions at the present may increase the mobility of uranium, flushing it out of surficial sediments, resulting in the low concentrations found in most sediments in southeast Arizona basins.

Correlation with Indoor-radon levels

A residential indoor-radon testing program was performed by the Arizona Radiation Regulatory Agency from 1987 to 1989. The U.S. Environmental Protection Agency has set a guideline indoor radon limit of 4 picocuries per liter (pCi/l), above which mitigation is recommended. Charcoal canister results from 22 homes in the lower San Pedro Valley show a mean indoor radon level of 1.26 pCi/l, only slightly higher than the average of about 1 pCi/l for all of Arizona. Ten homes (45%) registered at or below 1.0 pCi/l; None of the homes had radon levels higher than 4.0 pCi/l, compared with 5.4% of homes statewide. The highest reading from the valley was 2.4 pCi/l in the San Manuel area.

Studies by the U.S. Department of Energy have shown that water may be a significant source of radon in the home, with up to one third of indoor radon coming from water usage, particularly showers (U.S. Department of Energy, 1993). Information on the radon levels of water wells in the lower San Pedro valley are not available; a mean of 1148 pCi/l was reported for 32 wells sampled statewide (Duncan et al., 1993). The contribution to indoor radon from water in the lower San Pedro valley is undefined, but is probably low, considering the low levels of indoor radon measured in homes in the valley. The U.S. Environmental Protection Agency has recently proposed a limit of 300 pCi/l for radon in water.

RECONNAISSANCE OF URANIUM IN DRIPPING SPRING VALLEY

The Dripping Spring Valley is a small, isolated basin between Winkelman and Globe. The basin is bounded on the south by the Dripping Spring Mountains and on the north by the Mescal Mountains. A thick sequence of fluvial and lacustrine sediments (much like those in other basins in southeast Arizona) is exposed in the center of the valley and can be readily seen along State Highway 77. These lacustrine sediments include clay, silt, and marl, and at least four ash beds were reported by Scarborough (1975).

A reconnaissance survey of uranium levels was performed following the same procedures as for the San Pedro Valley. The sediments contained an average of 1.8 ppm uranium and no anomalous levels were encountered. Data on indoor radon levels for the few residences in the valley are not available.

CONCLUSION

The lower San Pedro Valley contains a few areas of slightly elevated uranium concentrations in diatomaceous basin-fill sediments, but no residences are located near these areas. Levels of uranium average 3.6 ppm for the entire valley, compared to an average level of 1.6 ppm determined in a statewide survey (Duncan and Spencer, 1993). Deleting the samples from the valley south of Mammoth yields an average for the rest of the valley of about 2.7 ppm, similar to the Safford and upper San Pedro Valleys (Harris, 1994; 1995) and similar to the estimated 3 ppm uranium average background level for the entire U.S. (Peake and Schuman, 1991)

Homes tested for indoor radon in the lower San Pedro valley have levels above the average for homes statewide (but are still below the EPA action level). However, considering the widespread distribution of lacustrine sediments in the basin, exposed and in the subsurface, the basin fill is a potentially significant source of indoor radon.

The possibility exists that other small or low-level uranium anomalies are present that were not

found during this survey. Further, the distribution of uranium in the subsurface basin-fill has not been determined.

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